Chapter 12 Pineapple Leaf Fibers and PALF-Reinforced Polymer Composites

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Abstract Pineapple leaf fibers (PALF) have long been known as textile materials in many countries. Despite being mechanically excellent and environmentally sound, PALF are the least-studied natural fibers, especially for reinforcing composites. This article presents a survey of research works carried out on PALF and PALF-reinforced composites. It reviews PALF extraction, fiber characterization, and PALF applications, modification of PALF, and manufacture and properties of PALF-reinforced composites. With increasing importance of pineapple and pineapple plantation area, value-added applications of PALF as reinforcing fibers in polymer composites must be developed in order to increase "resource potential" of pineapple and consequently energize the utilization of PALF.

Keywords Ananas comosus · PALF-reinforced composites · Pineapple leaf fibers

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12.1 Introduction

Polymer composites represent a class of versatile materials, each comprising a matrix binding a filling or reinforcing phase(s) resulting in a material with more desirable properties than those of the constituents taken individually (Mallick 1993). Matrices and synthetic fibers made from petroleum-based resources or produced using energy-intensive processes have been the standard constituents in making polymer composites until recently. Depletion of natural resources and serious environmental problems has spurred the search for renewable and more environment-friendly materials. The scientific community has since taken a serious relook at using natural plant fibers to reinforce composites like they used to be utilized during the early days.

Natural plant fibers have become popular among the scientific community as viable alternatives to glass fibers in fiber-reinforced plastics. These lignocellulosic fibers have lower densities, cost relatively lower to produce, consume lesser energies during production, pose no abrasion to machines and have no health risk when inhaled; in addition to being widely available, they are renewable and biodegradable and are CO_2 neutral (Wambua et al. 2003). To make it even more attractive, many of the fiber sources are at present agricultural wastes (Abdul Khalil et al. 2006).

One such fiber source known for a long time is pineapple leaves from which pineapple leaf fibers (PALF) may be extracted. Pineapple (*Ananas comosus*) is the third most important tropical fruit in the world after banana and citrus (Bartholomew et al. 2002). While reportedly having some of the highest mechanical properties (Bismarck et al. 2005), PALF are still the least-studied fibers, and pineapple leaves are mainly wasted through composting and burning. It is high time that value-added applications of PALF are identified as industrial significance and plantation area of pineapple in many countries increase (Anon 2008b). Development of PALF as reinforcing fibers in polymer composites shall improve the utilization of "resource potential" of pineapple (Benjamin and van Weenen 2000).

The properties of a composite are dictated by the intrinsic properties of the constituents which may be summarized as fiber architecture and fiber-matrix interface (Fowler et al. 2006). The reinforcing efficiency of natural fibers depends on their physical, chemical, and mechanical properties. Major shortcomings of natural plant fibers include fiber nonuniformity, property variation even between individual plants, low degradation temperature, low microbial resistance, and susceptibility to rotting. In addition to naturally occurring nonuniformity, fiber extraction and processing techniques also have major impacts on final fiber quality, not to mention fiber costs and yield (Munder et al. 2005).

Another serious drawback of using natural fibers with polymeric matrices is poor fiber-matrix interfacial adhesion, thus impairing composite mechanical and other properties. Optimization of interfacial adhesion between natural fibers and thermoplastic and thermoset matrices has been the focus of a large amount of research conducted during the past two decades (John and Anandjiwala 2008). Various means of modification applied on either the fibers or the matrix change not only the fiber–matrix interface, but also the fibers themselves.

12.2 Pineapple Leaf Fibers

PALF are obtained from the leaves of pineapple plant (*A. comosus*) from the family of Bromeliaceae (Anon 2008a) as shown in Fig. 12.1. Mwaikambo stated that pineapple plants are largely grown in tropical America, Africa, and Far-East Asian countries (Mwaikambo 2006). Thailand and the Philippines are the biggest pineapple fruit producers in the world (Anon 2008c). The Food and Agriculture Organization (FAO) has reported that most of the world pineapple fruit production in 2001 amounting to about 13.7 million tons of fresh fruits are produced in Asia (Anon 2008d). Pineapple leaves from the plantations are being wasted as they are cut after the fruits are harvested before being either composted or burnt. Ahmad et al. concluded that the practices of decomposing and burning the leaves in situ did not contribute to the improvement of plantation yield (Ahmed et al. 2002, 2004). Additionally, burning of these beneficial agricultural wastes causes environmental pollution.

12.2.1 PALF Applications

PALF have been traditionally used for making threads and textile fabrics in many countries. In the Philippines, PALF have been used for commercial products such as dresses, table linens, bags, mats, and other clothing items (Anon 2008b). PALF are also being utilized as textile materials in Indonesia, while works on using PALF for this application have just started in Malaysia (Mohamed et al. 2009). The use of



Fig. 12.1 Pineapple plant

chemically treated PALF in making industrial textiles, V-belt cord, conveyor belt cord, transmission cloth, air-bag tying cords, and shoe laces was reported in Basu et al. (2003). PALF can be blended with polyester fibers to replace jute for making needle-punched nonwovens for technical textiles (Hayavadana et al. 2003). The spinning, weaving, and processing of PALF were examined in Kumar et al. (1997), where the authors reported the use of PALF in making carpets and urged further studies on chemical processing, dyeing behavior, strength losses, and fiber degradation of PALF. PALF were blended with polyester fibers and silk, resulting in esthetically pleasing fabric and nonwovens (Anon 1992). The use of PALF to reinforce composites, the main focus of this review, shall be explained in the coming sections. Using PALF as textiles involves elaborate processes to increase the fiber fineness, blendability, and dyeing-ability. PALF dresses are known to be very costly understandably due to tedious manual processes involved. In contrast, the use of PALF as reinforcements in polymer composites may not involve such processes.

12.2.2 PALF Extraction

Pineapple leaves from which PALF are extracted are about 70–90 cm in length and 5–7.5 cm in width. Figure 12.2a shows the sword-shaped, dark green pineapple leaves, while Fig. 12.2b shows the extracted PALF bundles from the leaves. Manual extraction process can extract at least 3–4% of fibers from the leaves (Paul 1980). PALF are strong, white, and with a silky luster (Fig. 12.2c) and they are usually used to produce rope, broom, mat, nets, and cloths (Bhaduri et al. 1983).

Like other natural fibers, PALF vary in their properties according to species, geographical regions, age, locations in each plant, and weather conditions among others (Bismarck et al. 2005). There are many different species under the genus of *A. comosus*, while in the works on PALF and PALF-reinforced polymer composites reviewed, specific species of pineapple from which PALF were extracted were barely mentioned. Only in Bhaduri et al. (1983) the chemical compositions



Fig. 12.2 Pineapple leaf fibers (PALF) obtained from pineapple leaves

of PALF extracted from the leaves of three different varieties of the plant, viz. Giant Kew, Queen, and Mauritius were reported. It was reported in (Anon 1992) that various species of pineapple especially those with inedible fruits produced useful fibers for the textile industry. In light of species–property variability and lack of previous studies, Mohamed et al. (2009) carried out characterization of PALF extracted from three most popular species in Malaysia namely Queen (Moris Gajah), Smooth Cayenne (Sarawak), and Spanish (Josapine). They determined Josapine as the most appropriate species for PALF extraction considering potential fiber quantity, ease of extraction, fiber fineness, mechanical, and thermal properties.

Ways of obtaining PALF from the leaves have been crude and they continue to be hand-separated as the use of machines is generally slower than the manual process though it facilitates production processes (Mwaikambo 2006). Though simpler and more cost-effective, mechanical processes induce damage to natural fibers through breaking, scotching, and hackling actions leading to tensile strength of the elementary fibers to be only marginally higher than that of the technical fibers (Joffe et al. 2003). Manual extraction allows two types of fibers (Bartholomew et al. 2002) to be obtained from the leaves: 75 wt% of large vascular bundles present in the top lamina and 25 wt% of fine fiber strands in the bottom lamina (Mohamed 2010). Unlike bast fibers, PALF are readily available as distinct fiber bundles once they are separated from the leaf tissues. Large bundles are made up of loosely bound technical fibers which consist of elementary fibers with diameters less than 10 μ m.

A simple PALF separation technique using abrasive-combing in which large vascular bundles were pulled between #100 sandpapers was experimented in Mohamed et al. (2010a). Large bundles easily separated into two finer bundles reducing the mean PALF vascular bundle diameter by 50.3% (p = 0.01). Subsequent defibrillation using this technique was more difficult as experienced during manual separation of these bundles. The vascular bundles may therefore be visualized as being made up of two loosely bound parallel bundles, relatively speaking. The two bundles were made up of finer bundles not easily separated unless they were extracted with other mechanical or chemical means (Bismarck et al. 2005), and the smallest single bundles were expected to be those present in the bottom-most epidermal layer in the leaf.

Usually, plant fibers are retted to separate the fiber bundles from cementing materials resulting in finer bundles. Bacteria retting of PALF produced cleaner fibers with good tensile strength and reduced nonfibrous constituents as retting process converted much of the hemicellulose to α -cellulose (Palmario et al. 1976; Cueto et al. 1978). However, the problems associated with retting of natural fibers are limiting their competitiveness and range of applications (Hepworth et al. 2000). Hepworth et al. discovered that lack of retting did not significantly change the reinforcing capabilities of hemp fibers and the same may be reasonably assumed with PALF. Meanwhile they urged further study on possible degradation of unretted tissue over time after it has been embedded in resin for any serious effects on the useful life of the composites.

12.2.3 PALF Characterization

12.2.3.1 PALF Physical Properties

The average length of PALF bundles is approximately 1 m as reported in (Sharma 1981), while lower lengths (0.6–0.7 m) were noted in Mohamed et al. (2009). In Mohamed et al. (2010a) the authors measured PALF diameters in two perpendicular directions and, unlike the previous practice of assuming fiber circularity, used an ellipse as the chosen shape. They found an almost constant ratio of major to minor axis, hence the appropriateness of using ellipse to describe both large bundles and fine strands.

Majority of the previous works on PALF and PALF-reinforced composites utilized fine bundles with diameters less than 100 μ m. Mohamed et al. differentiated large vascular bundles and fine strands and determined that the former had a mean diameter of 241.9 μ m and was less varying, while the latter had a mean diameter of 72.7 μ m and a larger diameter variation as confirmed by visual observation (Mohamed et al. 2010a). They observed that PALF vascular bundles were present and extracted in one lamina, while fine fiber strands were present in a few bottom layers. The diameters of fine fiber strands were decreasing toward the bottom face of the leaf.

12.2.3.2 PALF Chemical Compositions and Structure

Table 12.1 shows PALF chemical composition obtained from previous studies. It can be seen that the content of cellulose was in the range of 67.12–82%; hemicelluloses 9.45–18.80%, hollocellulose 80–87.56%, lignin 4.4–15.4%, pectin 1.2–3%, fat and wax 3.2–4.2%, and ash 0.9–2.7%. Differing composition may be attributed to factors including source of fibers, age of fibers, climate conditions, and the process to obtain the fibers. Chemical composition, lignin distribution, and cell wall structure of PALF were analyzed in Abdul Khalil et al. (2006). Using transmission electron microscopy the authors confirmed that PALF cell wall structure

Chemical	Source							
composition (%)	Bhaduri et al. (1983)	Mohanty et al. (2000)	Abdul Khalil et al. (2006)	Saha et al. (1990)	Rowell and Han (2000)	Munirah et al. (2007)	Siregar et al. (2008)	
Cellulose	69.5	70-82	73.4	68.5	80-81	78.11	67.12-69.34	
Hemicellulose	-	_	_	18.80	16–19	9.45	_	
Hollocellulose	-	_	80.5	-	_	87.56	82.3-85.5	
Lignin	4.4	5-12.7	10.5	6.04	4.6-12	4.78	14.5-15.4	
Pectin	1.2	_	_	1.10	2–3	_	_	
Fat and wax	4.2	-	-	3.2	-	-	_	
Ash	2.7	_	2	0.9	_	_	1.21	
Extractive	_	_	5.5	-	_	-	3.83-0.97	

Table 12.1 Chemical compositions of PALF

may be described in terms of classical cell wall structure of a primary (P) and secondary (S1, S2, and S3) layers.

PALF have a multicellular structure with noncellulosic constituents like lignin and hemicelluloses holding the ultimate fibers together (Saha et al. 1990). The elementary unit of cellulose macromolecules is anhydro-D-glucosecopyranose, which contains three hydroxyls (–OH) per unit (Bledzki and Gassan 1999). These hydroxyls form intramolecular and intermolecular hydrogen bonds with each other as well as with hydroxyl groups from moist air (Mishra et al. 2001). Therefore, PALF are hydrophilic in nature and this feature affects the mechanical properties of PALF-reinforced composites (Abdul Khalil et al. 2007). The molecular chains of cellulose lie parallel and in three-dimensions in the crystalline region of PALF, while the other portions of the molecular chains are believed to lie in a less-ordered state in the amorphous regions (Mishra et al. 2004). Through X-ray diffraction (XRD) and thermogravimetric analyses, Mohamed et al. (2010a) concluded that the large PALF vascular bundles and fine fiber strands were chemically and structurally the same. XRD curves obtained resemble closely that of cellulose I (Nishino et al. 2003).

12.2.3.3 PALF Mechanical, Thermal, and Other Properties

Recently, the research community has shown growing interest in using PALF as to reinforce polymer composites due to their excellent mechanical properties compared to those of other natural fibers (Bismarck et al. 2005). Table 12.2 provides PALF mechanical properties obtained from previous studies. The tensile strength of PALF is in the range of 170–1,627 MPa, the tensile modulus is from 6.26 to 82.5 GPa, while the elongation at break 0.8–3.37%. Ultimately, high cellulose content and low microfibrillar angle of 14° (Bismarck et al. 2005) result in excellent PALF mechanical properties. Both flexural and torsional rigidity of PALF are comparable with jute fibers, while wet bundle strength of PALF decreases by 50% but the yarn strength increases by about 13% (Mishra et al. 2004).

Mechanical and	Source						
physical properties	Mohanty et al. (2000)	George et al. (1993)	Luo and Netravali (1995)	Arib et al. (2006)	Mohamed et al. (2009)		
Tensile strength (MPa)	413–1,627	170	445	126.6	293.08		
Young's modulus (GPa)	34.5-82.5	6.26	13.21	4.405	18.934		
Elongation at break (%)	1.6	0.8–1.6	3.37	2.2	1.41		
Diameter (µm)	20-80	5-30	_	_	105-300		
Density $(g \text{ cm}^{-3})$	-	1.44	1.36	1.07	_		
Moisture content (%)	11.8	-	_	-	-		
Microfibrillar angle (°)	14	12	_	_	-		

Table 12.2 Mechanical and physical properties of PALF

PALF tensile strength and Young's modulus decrease while elongation at break was not affected as fiber diameter increased (Mohamed et al. 2009, 2010a). Similar characteristics were also reported for another study on PALF (Mukherjee and Satyanarayana 1986). Unlike Mukherjee et al., Mohamed et al. differentiated large vascular bundles and fine fiber strands and tested them separately. The results suggested that it was not the absolute fiber diameter which dictated the mechanical properties but the number of individual fiber bundles making up larger bundles that did (Mohamed et al. 2010a). Fine PALF strands had high tensile strength and modulus and by linear regression gave strength and modulus values at 20 and 50 μ m of approximately 1800 MPa and 30 GPa and 750 MPa and 12 GPa, respectively. The strength and modulus of fine strands were very similar and within range of the values given in Bismarck et al. (2005) and Payae and Lopattananon (2009).

Normalizing mean strength of fine fiber strands at the mean diameter of large vascular bundles indicated that the vascular bundles were effectively stronger than the finer bundles (Mohamed 2010). This may be attributed to added strength of noncellulosic substances such as pectin and gums (Bel-Berger et al. 1999). Abrasive-combed PALF had 51.5% higher tensile strength and 48.6% greater moduli, while elongations at break did not differ much compared to those of nontreated PALF (p = 0.01) (Mohamed et al. 2010a). Linear regression analysis on data of fiber strands of diameters ranging 50–125 µm, however, suggested that the mean tensile strength was not much affected while modulus and elongation at break were reduced. This may be explained by possible introduction of defects on the defibrillated bundles through the action of the abrasives causing fraying and breaking of elementary fibers.

PALF mechanical properties are expected to decrease with temperature unless treated and modified. Most natural fibers lose their strength at about 160°C (George et al. 1996a). Removal of water present in natural fibers caused an increase in stiffness (Tomczak et al. 2007). The age of the pineapple plant is also expected to have some effects on fiber tensile strength and modulus, toughness, and elongation as found previously with sisal (Chand and Hashmi 1993). In Mohamed et al. (2009) thermal analysis of PALF from Josapine, Moris, and Sarawak cultivars gave curves similar in shape to that reported in George et al. (1996a). Major decomposition occurred at a temperature of approximately 340°C for Sarawak PALF and 320°C for Josapine and Moris PALF indicating higher thermal stability of Sarawak PALF. Interestingly, these decomposition temperatures were also higher than that reported on jute felt (Dash et al. 2002). In a more detailed study on Josapine PALF (Mohamed 2010), the authors concluded that large PALF vascular bundles and fine strands were thermally the same.

12.2.4 PALF Modifications

Good fiber-matrix interface is essential in the use of natural fibers as reinforcement in plastics to ensure performance and stability (Mwaikambo and Ansell 1999). The works on treatments and modifications of these hydrophilic lignocellulosic fibers were reviewed in John and Anandjiwala (2008). Various physical and chemical methods have been applied in order to overcome shortcomings of natural fibers like PALF which include low strength, fiber–matrix incompatibility, undesirable water uptake, and poor fiber–matrix adhesion. These include using chemical treatment (Joseph et al. 1996), surface modification (Mohanty et al. 2000; Danyadi et al. 2010), coupling agents (Araujo et al. 2008; Keener et al. 2004), and compatibilizing agent (Siregar et al. 2009).

12.2.4.1 PALF Pretreatments

The effects of simple pretreatments on the properties of PALF were studied in Mohamed (2010) and Mohamed et al. (2010a). The authors observed that washing PALF with water to remove leaf juices and dirt was found to be adequate but necessary in ensuring minimum fiber-matrix adhesion, while prolonged soaking was unnecessary unless retting with microorganisms (Palmario et al. 1976) is employed. Bleaching PALF with sodium hypochlorite (NaOCI) degraded the fibers mechanically and thermally as well as destroying PALF crystalline structure while PALF-vinyl ester adhesion was not improved. Hydrogen peroxide (H₂O₂) bleach improved PALF fineness by 5–6% but reduced the tensile strength by 40–45% (Ghosh et al. 1988). Bleached fibers when spun into yarns produced a more uniform and extensible but lower strength yarns than unbleached raw fibers.

12.2.4.2 Chemical Treatments

Alkali treatment or mercerization using sodium hydroxide (NaOH) is the most commonly used treatment for bleaching and cleaning the surface of natural fibers to produce high-quality fibers (Rahman and Khan 2007). Modifying natural fibers with alkali has greatly improved the mechanical properties of the resultant composites (Gomes et al. 2007; Van de Weyenberg et al. 2006). Additionally, alkali treatment leads to fiber fibrillation, thus increasing the effective surface area available for wetting by the matrix. Increasing fiber aspect ratio caused by reducing fiber diameter and producing a rough surface topography offer better fiber–matrix adhesion and increase in mechanical properties (de Albuquerque et al. 2000; George et al. 1998a). NaOH reacts with hydroxyl groups of the cementing materials in natural fibers and brings on the destruction of the cellular structure, thereby splitting the fibers into filaments (Cao et al. 2006).

Structural features and fracture morphology of raw and chemically treated PALF were studied by Saha et al. (1990) using scanning electron microscopy. Upon gradual removal of lignin and hemicellulose by alkali treatment, surface morphology progressively changed. Uniform sharing of the load in the ultimate fibers of raw PALF was observed, whereas treated fibers showed irregular fractures characterized by the slippage of ultimate fibers. Alkali treatment, nitration, dinitrophenylation, benzoylation, and benzoylation–acetylation of PALF have been reported

in Samal and Bhuyan (1994). Reduction in PALF percent moisture regain was significant upon dinitrophenylation and benzoylation–acetylation but low upon nitration, while higher mechanical strength was registered for all chemically modified fibers except for benzoylated PALF. NaOH treatment of PALF was also found to increase tensile modulus and strength by 25% and 19%, respectively, in addition to a 100% increase of PALF-epoxy interfacial shear strength (IFSS) (Payae and Lopattananon 2009). The authors observed even higher IFSS between epoxy-treated PALF and the matrix even though this treatment only increased PALF stiffness and not strength.

12.2.4.3 Chemical Coupling

Chemical coupling method utilized a compound to treat fiber surface to improve fiber-matrix interfacial adhesion through formation of a bridge of chemical bonds between the fiber and matrix. The interaction between silane coupling agents with natural fibers may be divided into four steps (a) hydrolysis, (b) self condensation, (c) adsorption, and (d) grafting (Xie et al. 2010). The effect of two different silane treatments namely, γ -(aminopropyl) trimethoxy silane (Z-6011) and γ -methacrylate propyl trimethoxy silane (Z-6030), on the mechanical properties of PALF-polycar-bonate composites was studied in Threepopnatkul et al. (2009). The modulus PALF/Z-60011-PC composites have higher modulus compared to PALF/Z-60011.

Graft copolymerization of acrylonitrile (AN) onto chemically modified PALF has been studied in Mohanty et al. (1996) using Ce(IV) and *N*-acetylglycine (NAG) combination as an initiator in the temperature range 40–60°C. Tripathy et al. studied the Cu(II)–IO₄⁻ initiated graft copolymerization of MMA from defatted PALF and investigated the effects of variation of time and temperature, concentration of Cu(II), KIO₄, and MMA, the amount of PALF, and also the effects of some inorganic salts and organic solvents on the percentage of graft yield (Tripathy et al. 1999). It has been shown that grafting improved the thermal stability of PALF. Graft copolymerization of AN and MMA onto PALF, using cerric ion as an initiator at varying concentrations of monomer, initiator, and mineral acid at a number of temperatures for various time intervals, was studied in Samal et al. (1994). Tensile data revealed a moderate increase in PALF tensile modulus when grafted with AN. Cyanoethylated PALF-graft-PAN samples had higher percentage of grafting but the increase in tensile modulus was not high.

12.3 PALF-Reinforced Polymer Composites

12.3.1 PALF-Thermoplastics Composites

Thermoplastics, especially commodity resins like polyethylene (PE) and polypropylene (PP), account for majority of the matrices used with PALF. Understandably, these commodity resins are inexpensive, widely available, and recyclable. Processing methods of natural fiber-reinforced thermoplastic composites include meltmixing and injection molding (George et al. 1996b; Abu-Sharkh and Hamid 2004; Threepopnatkul et al. 2009; Bledzki et al. 2010), sandwiching with compression molding (Luo and Netravali 1995; Arib et al. 2006; Mukhopadhyay and Srikanta 2008), melt mixing and compression molding (Antich et al. 2006; Kim et al. 2008), and extrusion and compression molding (Nair et al. 2001). Mixing natural fibers with thermoplastic matrices must be carried out at less than 200°C as most natural fibers lose their strength at temperature of 160°C (George et al. 1996a).

Geoge et al. carried out systematic studies on PALF-reinforced low-density polyethylene (LDPE) composites investigating the effects of PALF addition on composite viscoelastic properties (George et al. 1993), thermogravimetric and dynamic mechanical characteristics (George et al. 1996a), melt rheological behavior (George et al. 1996b), and electrical properties (George et al. 1997). They also evaluated the environmental effects on the properties of PALF-LDPE (George et al. 1998b), stress relaxation behavior of short PALF-LDPE composites (George et al. 1998c), and the effects of strain rate and temperature on the tensile failure of PALF-LDPE composites (George et al. 1998c). In studying short PALF-LDPE composites the authors determined the optimum parameters for melt mixing process to be 6 min of mixing at rotor speed of 60 rpm and mixing temperature of 130°C (George et al. 1995). Fiber length of 6 mm was the most suitable length for enhanced properties in solution mixing.

Incorporating PALF in LDPE in increasing amount caused increased water uptake and decreased mechanical properties (George et al. 1998b), increased dynamic storage and loss modulus (George et al. 1993, 1996a), lowered degradation temperature (George et al. 1996a), increased viscosity (George et al. 1996b), increase in dielectric constants and decreased volumetric resistivity and dielectric dissipation factor values (George et al. 1997), decreased rate of relaxation (George et al. 1998c), and better strength retention at higher temperature (George et al. 1999). Fiber treatments using NaOH, silane A-172, isocyanate (PMPPIC), and benzoyl peroxide carried out in the above studies improved fiber–matrix adhesion, and in many instances enhanced the positive gains or reduced the undesirable properties as a result of PALF addition.

The effect of fiber loading on mechanical properties of unidirectional PALFreinforced PP composites produced by compression molding was investigated in Arib et al. (2006). Tensile modulus and strength increased with increasing fiber volume fraction with maximum values obtained at 10.8% PALF before decreasing slightly at 16.2%. Increases in flexural modulus and stress were relatively low attributed to fiber–fiber interaction, voids, and dispersion problems. Injectionmolded polycarbonate (PC) reinforced with PALF pretreated with NaOH and modified with two types of silane was studied in Threepopnatkul et al. (2009). The authors observed that composite mechanical properties were better than neat PC, while silanized composites have lower mechanical property than NaOH-treated composites due to the formation of flexible polysiloxane. Siregar and Sapuan (2009) studied the mechanical properties of PALF-reinforced high impact polystyrene (HIPS) composites utilizing ground PALF fibers of 10–40, 40–60, and 60–80 meshes. They observed that adding PALF of various sizes at different fiber loadings resulted in increases in composite tensile and flexural moduli and hardness. However, tensile and flexural strengths and notched and un-notched impact strength of the composites decreased. Increase in fiber content caused increase in the moduli while strength decreased.

12.3.2 PALF Thermoset Polymer Composites

Polyester, epoxy, phenol formaldehyde (PF), and vinyl ester are typical thermosets used in natural fiber-reinforced composites, with polyester being the most widely used (Sanadi et al. 1986). Compression molding is a common and convenient method to make these composites regardless of fiber length (Mishra et al. 2004), though in the case of PALF-reinforced thermoset composites, no indication of high consolidation pressure had been observed. Polyester composites reinforced with unidirectional natural fibers including PALF were studied in Pavithran et al. (1987), and the composite toughness of the composites was observed to increase with the fiber microfibrillar angle. The influence of fiber length, fiber loading, and coupling agents on tensile, flexural, and impact properties of PALF-reinforced polyester composites was investigated in Uma Devi et al. (1997). With optimum fiber length of 30 mm, composite tensile strength, Young's modulus, flexural strength, and impact strength increased linearly with fiber weight fraction. In the case of flexural strength, however, a leveling off beyond 30% was observed. This trend was also observed in another study of PALF-polyester composites (Mishra et al. 2001). 30 wt% of PALF seemed to result in maximum tensile, flexural, and impact strengths with corresponding decreases thereafter. In another study, thermal conductivity of PALF/PF composites decreased with the increasing fiber content (Mangal et al. 2003). The addition of varying PALF content (10-50%) to reinforce PF composites cannot improve composite thermal conductivity due to low PALF thermal conductivity.

Recently Payae and Lopattananon (2009) studied the use of epoxy as the matrix for PALF even though they stopped short of studying PALF-reinforced epoxy composites. The use of PALF to reinforce vinyl ester was studied by Mohamed (2010) and Mohamed et al. (2010b). Vinyl esters are in between polyester and epoxy in terms of cost, ease of processing, and performance (Li 1998) and are tough, resilient, and less susceptible to water degradation (Joffe et al. 2003). Liquid compression molding of PALF with vinyl ester was tried in Mohamed et al. (2010b). It was observed that the reinforcing capability of PALF stored for 6 months under hot and humid ambient conditions was not degraded (Mohamed 2010). The authors noted that PALF may be utilized at random from different parts of the leaves without affecting composite flexural properties. Utilizing large vascular bundles and fine fiber strands separately at 20 wt% and low consolidating pressure suggested that fiber diameter hardly affects flexural strength and modulus.

Property	Unit	PALF-polyester ^a	PALF-polyester ^b	PALF–vinyl ester ^c	PALF-vinyl ester ^d
Untreated PALF					
Tensile strength	(MPa)	52.9	42	73.2	_
Flexural strength	(MPa)	80.2	86.28	98.9	122
Impact strength Treated PALF	(kJ m ⁻²)	24.2	-	19.2	105 ^e
Tensile strength	(MPa)	73.5 ^f	43.6 ^g	-	_
Flexural strength	(MPa)	85.6 ^f	117 ^g	_	_
Impact strength	(kJ/m^2)	-	102.4 ^{e,f}	_	_

Table 12.3 Selected tensile, flexural, and impact strengths of PALF-reinforced thermoset composites

Values either categorically stated or inferred from the figures.

^aRandomly oriented 30 mm PALF (30 wt%) (Uma Devi et al. 1997)

^bRandom nonwoven PALF mat (30 wt%) (Mishra et al. 2001)

^cCompression molded, randomly oriented nonwoven PALF mat (28.4 wt%) (Mohamed et al. 2010b)

^dHand laid, unidirectional PALF (20 wt%) (Mohamed 2010)

^e(J/m)

^fSilane A-172

gCyanoethylated PALF

Using fine fiber strands, however, significantly improved the toughness of PALF–vinyl ester composite due to greater number of interfaces generated. Composites reinforced with abrasive-combed PALF though finer than large vascular bundles were less tough than those using coarser bundles due to defects present on them, thereby highlighting the importance of fiber integrity.

Effects of fiber treatments and modifications on the properties of PALF-thermoset composites were also studied by the above workers. The effects of dewaxing, alkali treatment, cyanoethylation, and grafting of AN onto dewaxed PALF on composite mechanical properties were studied in Mishra et al. (2001). Amongst all the modifications, 10% AN-grafted PALF composites exhibited maximum tensile strength, whereas cyanoethylated PALF composite exhibited better flexural and impact strengths. In another study, a significant increase in the strength of the composites was observed using PALF treated with silane A-172 (Uma Devi et al. 1997). Table 12.3 provides a summary of selected tensile, flexural, and impact strengths of PALF-reinforced thermoset composites.

12.3.3 Other Matrices Reinforced with PALF

Mechanical properties of PALF-reinforced natural rubber composites were studied in Bhattacharya et al. (1986) in which PALF addition increased hardness, compression set, tear resistance, and Mooney viscosity. However, elongation at break, mill shrinkage, and Mooney scorch time decreased. The use of resorcinol, hexamethylenetetramine, and silica is essential for PALF-rubber adhesion. PALF-reinforced natural rubbers filled with carbon black have high hardness, low elongation, moderate tensile strength, and moderate flex resistance.

Ideally, natural polymer matrices are used with PALF to make totally green composites. Currently, performance limitations and high cost of biopolymers are major barriers for their widespread acceptance as substitutes to traditional petroleumbased polymers (Mohanty et al. 2005). The low value interfacial shear stress of PALF/poly(hydroxybutyrate-co-valerate) (PHBV) reported in Luo and Netravali (1999) gave a clue of the performance limitations of biopolymers. Many of them are still being developed using elaborate chemical reactions in laboratories. In Luo and Netravali (1995) the authors utilized PALF to reinforce PHBV composites prepared by sandwiching three layers of PALF fibers between four layers of PHBV films and compression molding them with temperature and pressure of 180°C and 140 MPa respectively. Their study showed that adding 20 wt% unidirectional PALF enhanced the composite tensile and flexural strength by 75% and 32%, respectively. Tensile and flexural strengths increased by 100% and 60%, respectively, with 30 wt% PALF addition. Other matrices used with PALF include biodegradable polymers such as petroleum-based polyester amide (BAK 1095) (Mishra et al. 2002) and cornbased poly(lactic), PLA (Liu et al. 2005).

12.3.4 Hybrid PALF-Reinforced Composites

Hybrid biocomposites are formed by incorporating two or more fibers into a single matrix resulting in an apparent synergistic improvement in composite properties and favorable balance between the inherent advantages and disadvantages of individual components (John et al. 2009). Mishra et al. (2003) evaluated the mechanical properties of PALF/glass fiber-reinforced polyester composites and observed improvement in tensile, flexural, and impact strengths of the composites through incorporation of a small amount of glass fibers indicating positive hybrid effects. An optimum glass fiber loading of 8.6 wt% was noted. The PALF/glass fiber-reinforced polyester composites also showed significant reduction in water uptake compared to unhybridized samples. Thermophysical properties of PALF/ glass fiber-reinforced polyester composites were also studied in Idicula et al. (2006). Varying hybrid effects may be calculated for the mean values of thermal conductivity, diffusivity, specific heat, and density obtained.

12.4 Conclusions

PALF are environmentally sound and mechanically excellent materials to reinforce polymer composites for making industrial goods. As the industrial importance and plantation area of pineapple increase, it makes environmental and economical sense to energize the research and development to enhance PALF as reinforcing fibers. Developmental works must be carried out to mechanize PALF extraction and

separation techniques in order to cost-effectively produce good quality fibers. Polymer composite applications using PALF as reinforcing or filling phase must be quickly developed and commercialized in order to increase pineapple "resource potential" and consequently push the development and utilization of PALF and subsequently reduce wastage and environmental degradation. Systematic studies must be carried out to determine the most influential factors dictating the quality of PALF-reinforced polymer composites and allow optimization to be further carried out. Proper balance of performance and costs may then be achieved with the final aim to contribute positively toward the environment and well-being of the people. Hybridization of PALF with other biofibers or synthetic fibers in order to obtain "net positive hybrid effect" may still be explored. The use of biopolymers or blends of matrices in conjunction with PALF must also be given adequate attention.

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